



Effect of Layer-Graded Bond Coats on Edge Stress Concentration and Oxidation Behavior of Thermal Barrier Coatings

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EFFECT OF LAYER-GRADED BOND COATS ON EDGE STRESS CONCENTRATION AND OXIDATION BEHAVIOR OF THERMAL BARRIER COATINGS

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ABSTRACT

Thermal barrier coating (TBC) durability is closely related to design, processing and microstructure of the coating systems. Two important issues that must be considered during the design of a thermal barrier coating are thermal expansion and modulus mismatch between the substrate and the ceramic layer, and substrate oxidation. In many cases, both of these issues may be best addressed through the selection of an appropriate bond coat system. In this study, a low thermal expansion and layer-graded bond coat system, that consists of plasma-sprayed FeCoNiCrAl and FeCrAlY coatings and a high velocity oxyfuel (HVOF) sprayed FeCrAlY coating, is developed to minimize the thermal stresses and provide oxidation resistance. The thermal expansion and oxidation behavior of the coating system are also characterized, and the strain isolation effect of the bond coat system is analyzed using the finite element method (FEM). Experiments and finite element results show that the layer-graded bond coat system possesses lower interfacial stresses, better strain isolation and excellent oxidation resistance, thus significantly improving the coating performance and durability.

I. INTRODUCTION

Ceramic thermal barrier coatings (TBC) have been developed for advanced gas turbine ^[1, 2] and diesel engine applications ^[3-5] to improve engine durability and fuel efficiency. However, durability issues of these thermal barrier coatings under high temperature cyclic conditions are still of major concern. The coating delamination failure is closely related to thermal stresses in the coating systems, and oxidation of bond coats and substrate ^[6-9]. Coating shrinkage and through-thickness cracking resulting from ceramic sintering and creep at high temperatures ^[10], will further accelerate the coating failure process due to edge stress concentration effects along the coating interfaces. The coating reliability can be greatly improved through the development of an appropriate bond coat

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system. The bond coat system should not only be oxidation resistant, but also be tailored to grade the thermal expansion differences (provide strain isolation for the ceramic coating) and protect the substrate from oxidation. Thus, the stress arising from thermal expansion can be minimized and interfacial strength can be maintained. The strain isolation provided by a graded bond coat is especially beneficial for coating edges that exist due to component geometry, or due to through-thickness cracking resulting from ceramic sintering and creep at high temperatures. This layer-graded bond coat concept was proposed in earlier work where thermal fatigue testing of graded bond coat systems showed improved performance^[11]. In the present study, the thermal expansion and oxidation behavior of a low thermal expansion, three-layer-graded bond coat system are investigated. This coating system is comprised of plasma-sprayed FeCoNiCrAl, and FeCrAlY coatings and a high velocity oxyfuel (HVOF) sprayed FeCrAlY coating, as shown in Figure 1. The strain isolation effect of the bond coat system and the interfacial elastic stresses are analyzed using dilatometry and the finite element method (FEM). The influence of bond coat thickness on coating interface delaminations is evaluated based on thermal expansion behavior of the TBC system and the ceramic/metal interface microstructure characterization after thermal cycling experiments.

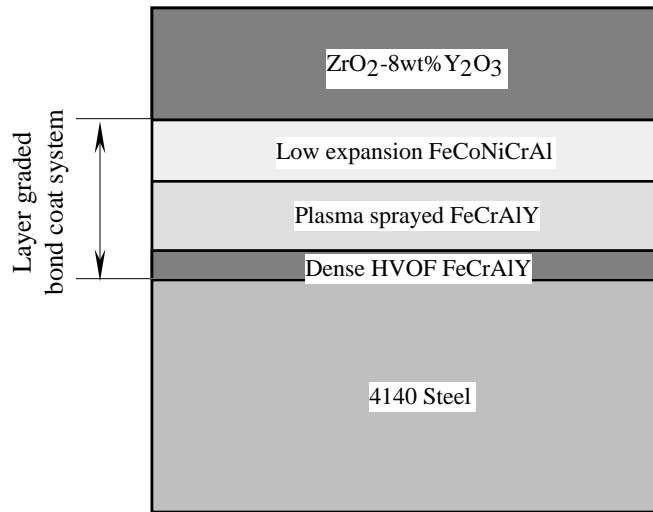


Fig. 1 Schematic diagram showing a layer-graded thermal barrier coating system that consists of a plasma-sprayed $\text{ZrO}_2\text{-8\%Y}_2\text{O}_3$ top coat, and a plasma-sprayed FeCoNiCrAl, FeCrAlY and thin HVOF sprayed FeCrAlY bond coat system.

II. EXPERIMENTAL METHODS AND MATERIALS

The oxidation kinetics of air plasma-sprayed Fe-28Co-24Ni-5Cr-4Al and Fe-25Cr-5Al-0.5Y bond coats were determined by thermogravimetric analysis (TGA) in flowing air, using the free-standing bond coat coupons (specimen dimension $25.4 \times 12.7 \times 1$ mm). The oxidation kinetics of 4140 steel were determined by TGA for the uncoated 4140 specimens, and by measurements of the oxide scale thicknesses from the cross-sections of the coated diameter 12.7 mm disk specimens, respectively.

Thermal expansion experiments were carried out on free-standing ceramic and bond coat materials in air using a dilatometer system. Thermal expansion response of the TBC system was conducted on the ceramic coatings attached to the substrate in high purity argon using dilatometry, as shown in Figure 2. The specimen length was 25.4 mm, and a platinum standard specimen was used as reference. The thickness of the ceramic coating was 1.5 mm, and thicknesses of the bond coats were chosen as 0.1-0.5 mm. The 4140 steel substrate was 12.7 mm in thickness to ensure no significant bending during the experiments.

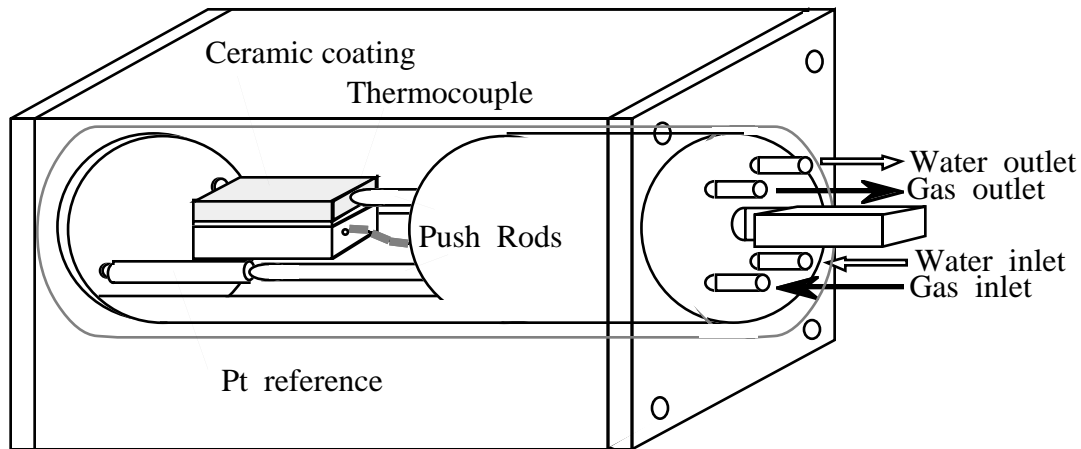
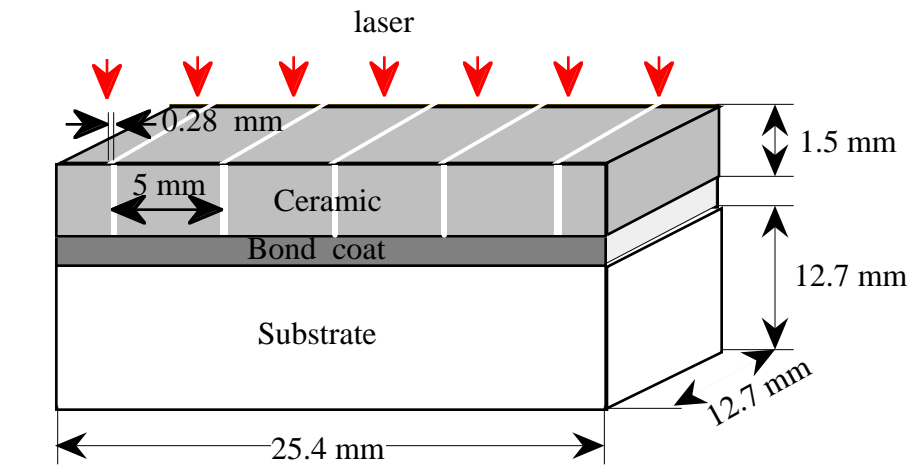
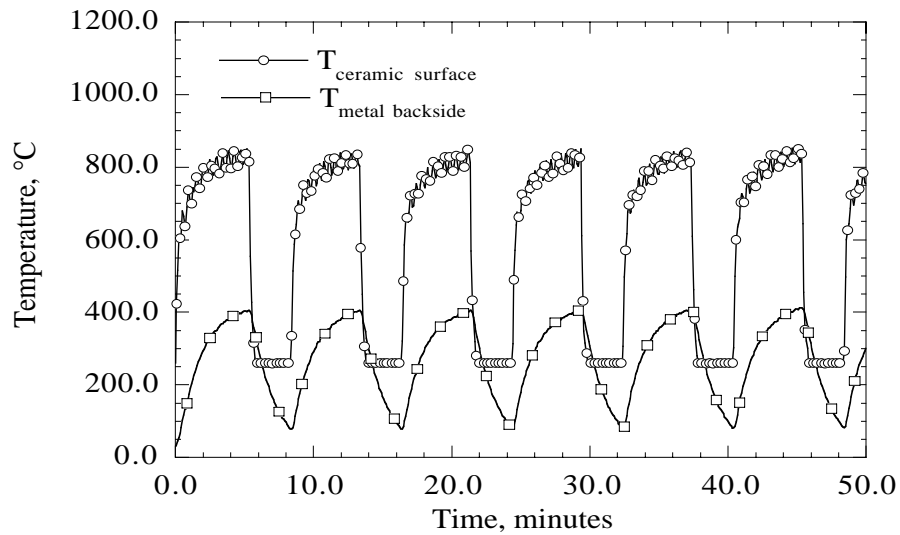


Fig. 2 The dilatometer system used for the measurement of thermal expansion behavior of thermal barrier coating systems.

In order to investigate the bond coat type on edge crack initiation and propagation, segmented thermal barrier coating systems (1.5 mm thick ceramic coating on $25.4 \times 12.7 \times 12.7$ mm steel substrate with various bond coat thicknesses) were tested up to 50 cycles (5 minute heating and 3 minute cooling) under laser thermal gradient cyclic conditions. Before each test, the ceramic coating was segmented with a diamond saw, so through-thickness, parallel notches were formed in the ceramic coating. The coating segment length (the spacing between notches) and the notch width were approximately 5 mm and 0.28 mm, respectively, as shown in Figure 3 (a). These notch edges near the ceramic/bond coat interface were used to simulate the coating sintering segmentation induced edges. The segmented TBC specimens were then thermally cycled using a rotating laser rig, with the uniform laser beam heating the entire ceramic coating surface. The substrate backside air cooling was used to establish the temperature gradients across the coating system. During these tests, the maximum ceramic surface and back side metal temperatures were maintained at about 850°C and 400°C, respectively. The typical heating and cooling temperature profiles are illustrated in Figure 3 (b).



(a)



(b)

Fig. 3 Laser thermal gradient cycling experiments of thermal barrier coatings. (a) Segmented ceramic coating specimen configuration for laser testing; (b) Typical heating and cooling cycles for the thermal barrier coating systems under laser thermal cycling conditions.

III. STRESS ANALYSIS PROCEDURE

The analytical procedure implemented in this study is the Finite Element Methods (FEM). Two-dimensional FEM meshes were generated for various specimen lengths and various bond coat systems. A typical FEM mesh is shown in Figure 4 (a) for a half TBC specimen length of 12.7 mm. The metal substrate is 12.7 mm high and the ceramic coating is around 1.5 mm. Two bond coat layers are sandwiched in between. The bond coat layer thicknesses were in the range of 0.1 to 0.5 mm. The mesh consisted of a total of 1024 eight-noded quadrilateral elements with 3201 nodes. The element sizes decreased approaching the free edge (the right hand-side) as well as the sandwiched bond coats to capture the sharp stress gradients at these locations. A total of 32 unequal elements span the length of the specimen with a higher element density at the free edge. While the steel substrate is meshed by 16x32 layers, the ceramic coating is layer by 8x32 elements since the ceramic coating is much smaller than the steel substrate. The two bond coat layers have 4x32 elements each. The use of only four layers for each bond coat is deemed appropriate given the parabolic variation in displacement and stresses formulated in the eight-noded element used.

The numerical analyses were implemented in the ABAQUS general purpose finite code ^[12]. Only elastic stress analyses were considered in this study. The material properties assumed in the analyses are given in Table 1. In this analysis, the stress free temperature is assumed to be at room temperature. The effect of the residual stress built up from the plasma spraying of the bond coat systems is ignored. For constant temperature simulations only a stress analysis is required, but for the through thickness simulations, a steady state heat transfer analysis is required to determine the nodal temperature followed by the elastic stress analysis. A typical mesh of the segmented TBC specimen is shown in Figure 4 (b).

Table 1 Physical and mechanical properties of the thermal barrier coating systems used in the FEM stress calculations ^[11]

Materials Properties	Plasma Sprayed ZrO ₂ -8wt% Y ₂ O ₃	4140 steel	Plasma Sprayed FeCrAlY	Plasma Sprayed FeCoNiCrAl
Thermal Conductivity k , W/m K	0.9	46.7	11	11
Thermal Expansion Coefficient α , m/m K	10.8×10^{-6}	14.2×10^{-6}	12.5×10^{-6}	11.0×10^{-6}
Density ρ , kg/m ³	5236	7850	6500	6500
Heat Capacity c , J/kg K	582	456.4	575	575
Young's modulus E, GPa	27.6	207	137.9	100

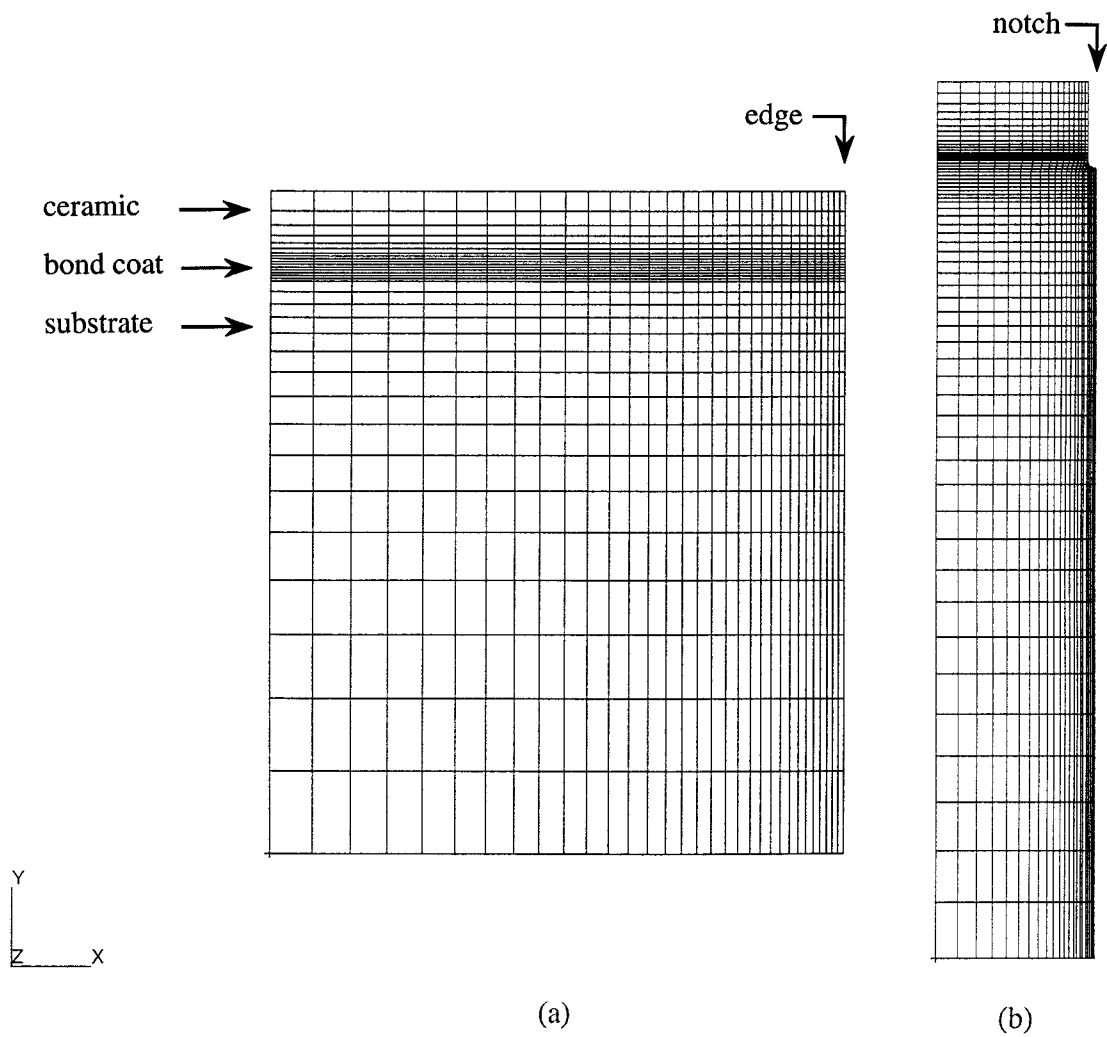


Fig. 4 Typical FEM meshes used in the stress analysis. (a) A FEM mesh for a half TBC specimen length of 12.7 mm. (b) A FEM mesh for a half segment TBC specimen length of 2.5 mm.

IV. RESULTS AND DISCUSSION

The air plasma-sprayed bond coats exhibited a complicated transient oxidation behavior due to its relatively high porosity, as has been reported previously ^[7]. Figure 5 shows the Arrhenius plots of apparent oxidation parabolic rate constants of the plasma-sprayed FeCoCrNiAl and FeCrAlY bond coats. As compared to the commonly used Ni-36Cr-5Al-Y bond coat, the FeCrAlY bond coat showed a faster oxidation rate mainly at the lower temperatures. The FeCoCrNiAl showed the fastest oxidation rate. X-ray diffraction results suggested that the plasma sprayed FeCrAlY and FeCoCrNiAl bond coats sometimes can form non-protective Fe-containing oxides at low oxidation temperatures. Air plasma-sprayed FeCrAlY bond coat could not effectively prevent the 4140 steel substrates from oxidation due to its relatively high porosity in the coating ^[7]. However, the denser HVOF sprayed FeCrAlY bond coat provided a higher bond strength, and showed better oxidation protection for the substrate. Figure 6 shows typical microstructures of the plasma-sprayed FeCrAlY bond coat after oxidation at 800°C for 50 hours.

Figure 7 shows the experimentally measured thermal expansion behavior of the free-standing ceramic coating, bond coat and steel substrate materials used in the TBC systems. As have been determined in an early study ^[7], the 4140 steel and ZrO₂-8% Y₂O₃ ceramic possess the highest and the lowest thermal expansion coefficients, respectively, while the air-plasma-sprayed FeCrAlY bond coat shows an intermediate thermal expansion coefficient. The low thermal expansion bond coat and FeCrNiCoAl has a similar coefficient of thermal expansion to the ceramic coating at temperatures below 600°C.

Due to the thermal expansion mismatch in the ceramic coating and substrate, thermal stresses are generated in the coating system under thermal cycling conditions. Stress concentrations near the coating edges due to the temperature change can lead to coating cracking and delamination along the ceramic/metal interfaces. Figure 8 shows typical stress distributions predicted by FEM in thermal barrier coating systems with the finite length specimen geometries under the condition of uniform temperature variation from 25 to 700°C. The stress distributions near the coating edges in this uniform heating case are characterized by the essentially very low normal in-plane stress component σ_{xx} , the high normal tensile stress component σ_{yy} perpendicular to the interfaces and shear stress component σ_{xy} along the ceramic/bond coat and the bond coat/substrate interfaces. The tensile strength of the ceramic coatings varies from 30 to 50 MPa, the large tensile stress component σ_{yy} developed during the heating cycle near the coating edges can be an important factor for coating delamination. The shear strength of the ceramic coating parallel to the bond coat is approximately 10 to 14 MPa ^[13], delamination cracks can also be easily initiated at coating edges near the ceramic/bond coat interface where the highest shear stress σ_{xy} is expected. However, as will be discussed later, the coating shear delamination becomes a predominant mechanism under thermal gradient conditions, because the tensile stress component σ_{yy} near the coating edge can be significantly lowered or even be slightly compressive for the non-uniform heating case. Oxidation of the bond coat and substrate will complicate this process, and in general facilitate the coating delamination.

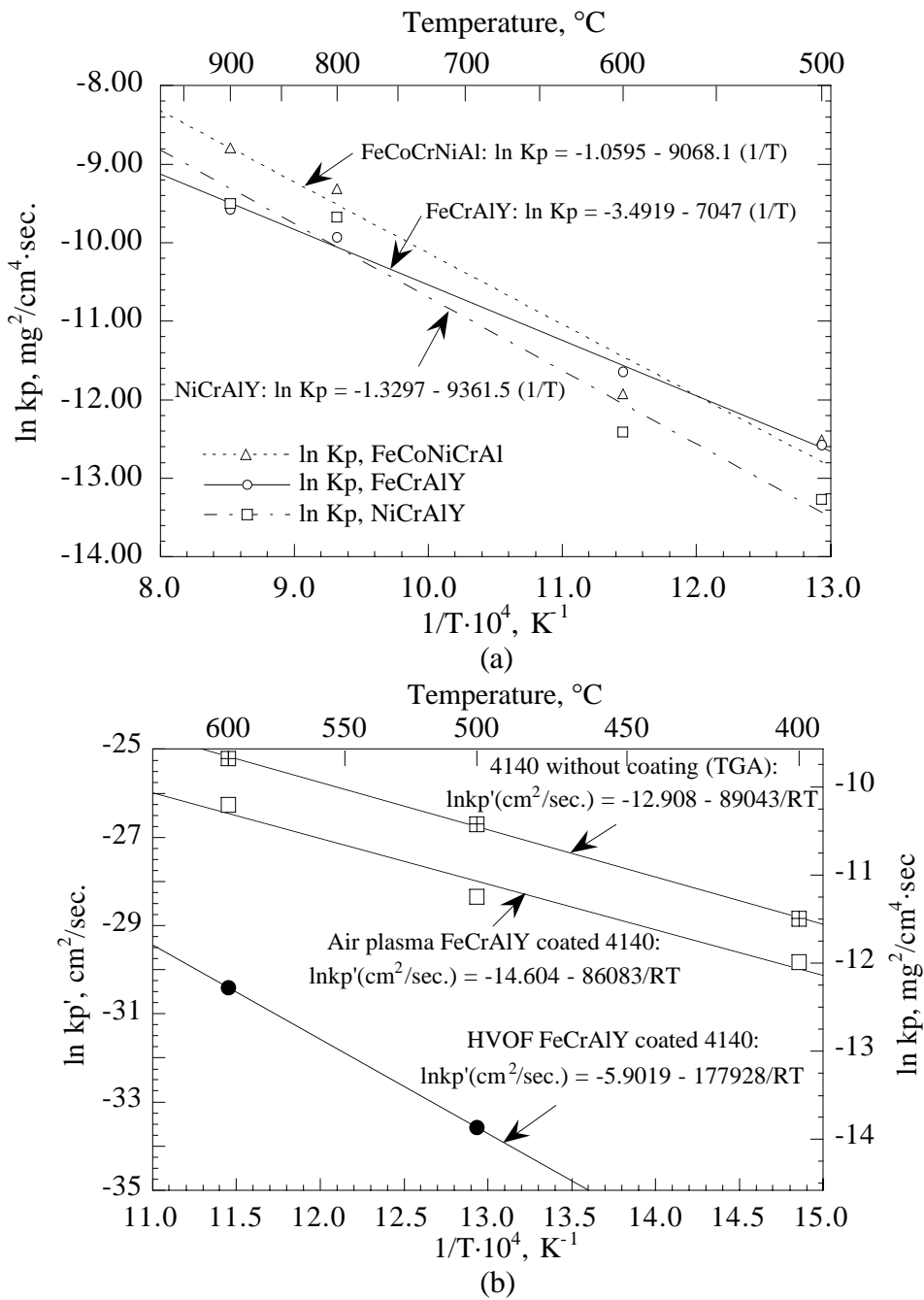
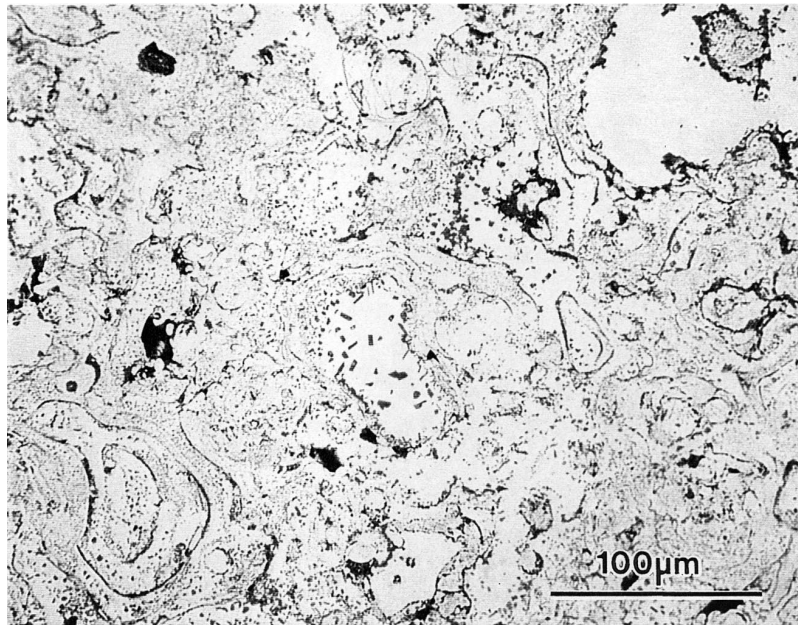
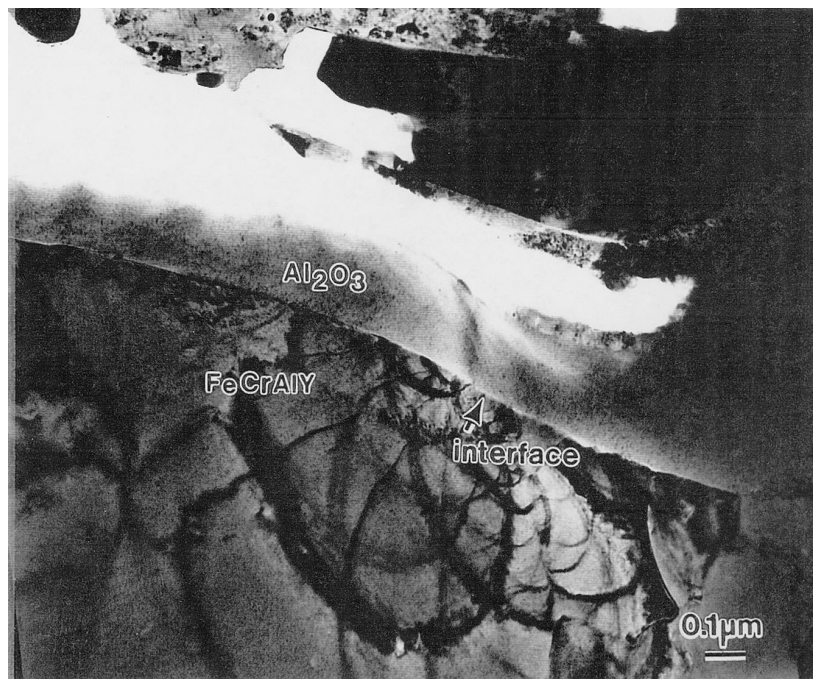


Fig. 5 Arrhenius plots of the parabolic rate constants of the plasma sprayed bond coats and the 4140 steel substrate. (a) $\ln k_p$ (in $\text{mg}^2/\text{cm}^4 \cdot \text{sec.}$) - $1/T$ relations for the free standing FeCoNiCrAl, FeCrAlY and NiCrAlY specimens by TGA measurements; (b) $\ln k_p'$ (in $\text{cm}^2/\text{sec.}$) - $1/T$ relation for 4140 steel specimens with and without FeCrAlY bond coat by scale thickness measurements.



(a)



(b)

Fig. 6 Typical microstructures of the plasma-sprayed FeCrAlY bond coat after oxidation at 800°C. (a) Optical micrograph of the coating section parallel to the ceramic/bond coat interface; (b) Transmission electron micrograph of the coating showing the Al_2O_3 scales grown on the FeCrAlY grains.

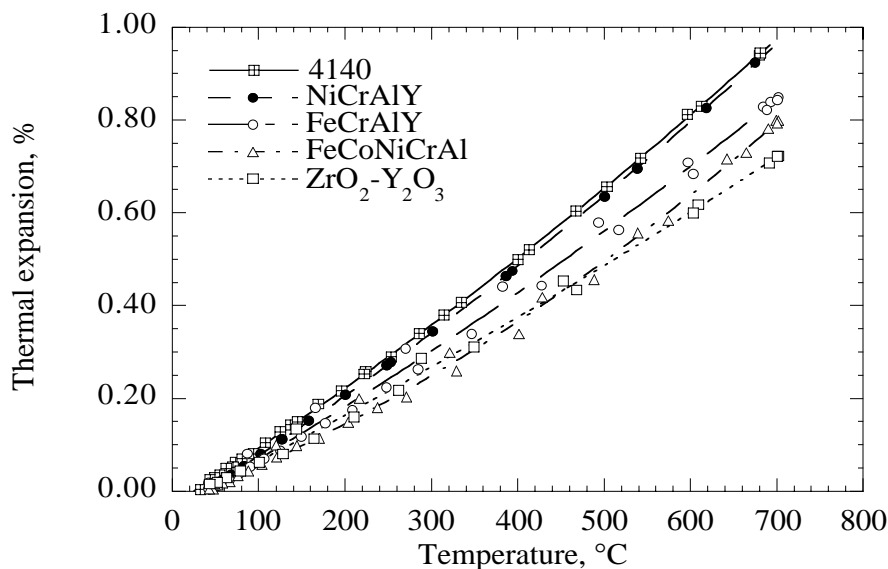


Fig. 7 Experimentally measured thermal expansion behavior of the free-standing ceramic coating, bond coat and substrate materials.

Oxidation resistant and thermal expansion-graded bond coat systems can help minimize shear crack initiation and propagation by improving the stress distributions near the interfaces and the interface adhesion. The FEM calculation results shown in Figure 8 demonstrate that the two layer-graded bond coat system can reduce the both shear and tensile stress concentrations near the ceramic/metal interface, and shift the peak stress away from the edge, compared with no bond coat or only the FeCrAlY bond coat case. In addition, with the two layer low expansion bond coat system, the overall stress levels are lowered and the high stress region is shifted to the bond coat/substrate interface.

The stress concentrations in TBC systems are also closely related to the bond coat thickness and specimen size. Figure 9 illustrates the influence of the bond coat thickness and the specimen length on shear stress distribution. For specimens of various lengths, the edge stress concentration increases with decreasing total bond coat thickness. In addition, the high shear stress regions occupy a larger portion of the ceramic/metal interface area for a shorter segment length, thus increasing the trend to delaminate the coating by the shear mechanism near the interface.

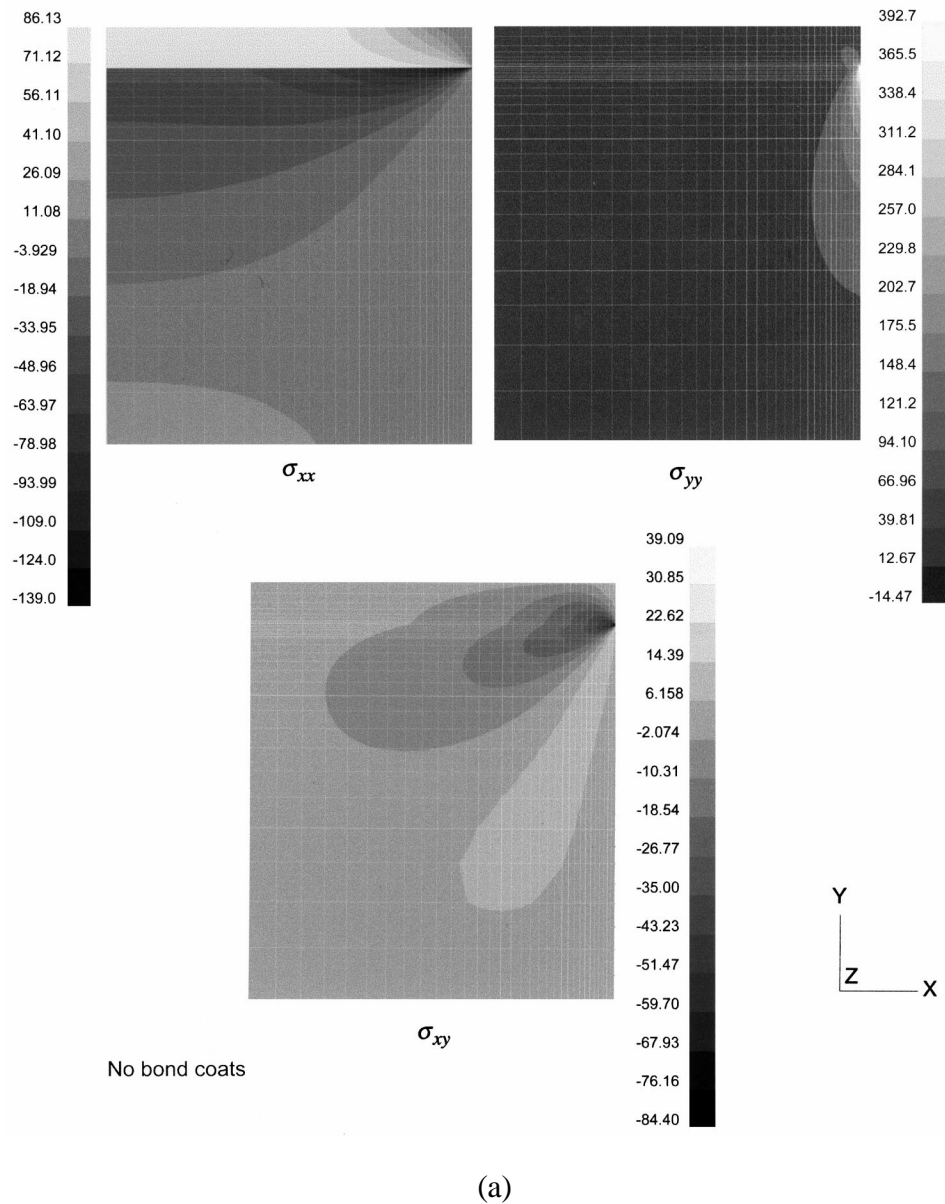


Fig. 8 FEM stress distributions in thermal barrier coating systems under the condition of uniform temperature variation from 25 to 700°C. (a) Normal and shear stress distributions of a coating system without bond coats; (b) Normal and shear stress distributions of a coating system with two layer bond coats; (c) Influence of the bond coat type on edge shear stress distributions in the ceramic coating at the ceramic/bond coat interface.

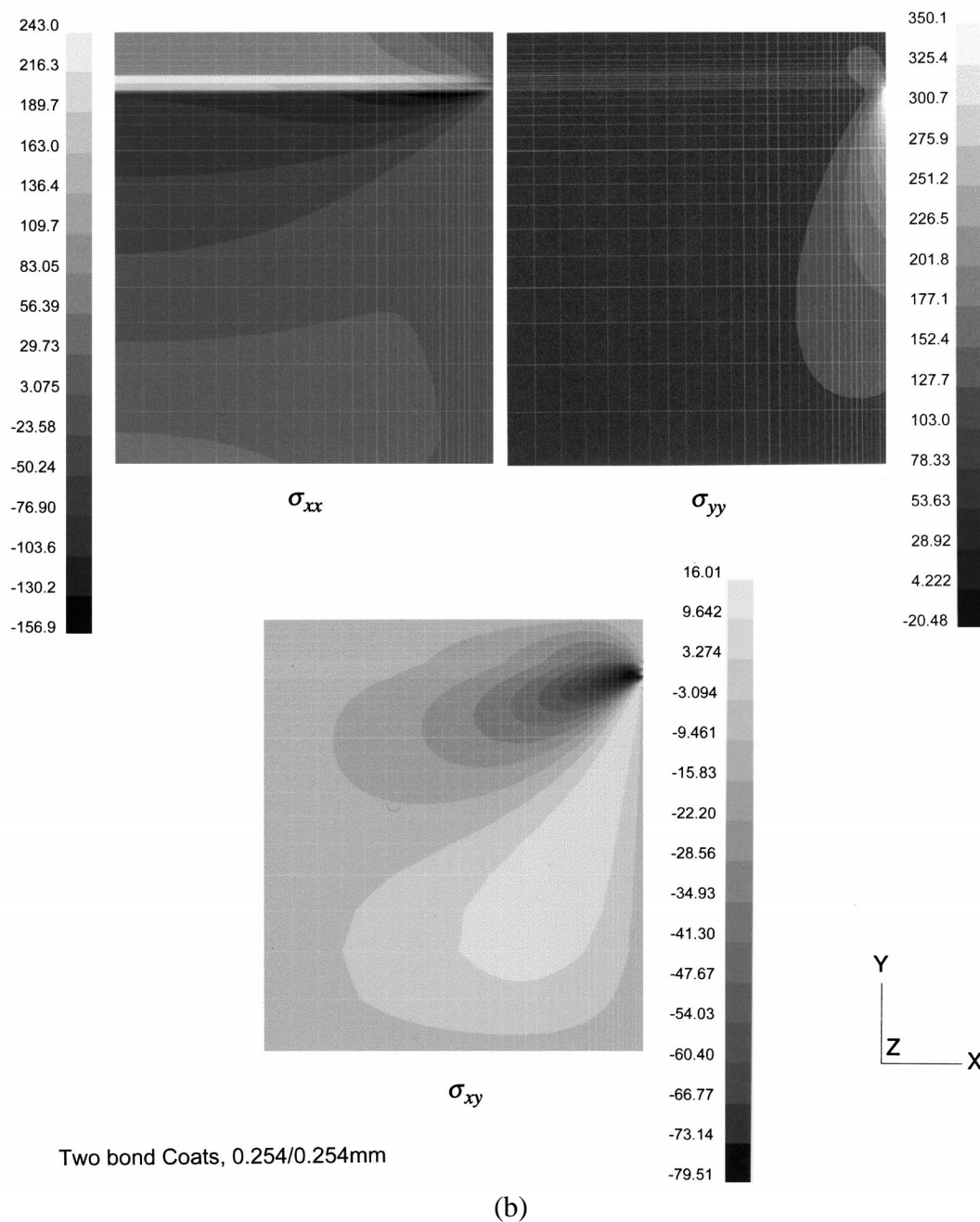


Fig. 8 (Continued) FEM stress distributions in thermal barrier coating systems under the condition of uniform temperature variation from 25 to 700°C. (a) Normal and shear stress distributions of a coating system without bond coats; (b) Normal and shear stress distributions of a coating system with two layer bond coats; (c) Influence of the bond coat type on edge shear stress distributions in the ceramic coating at the ceramic/bond coat interface.

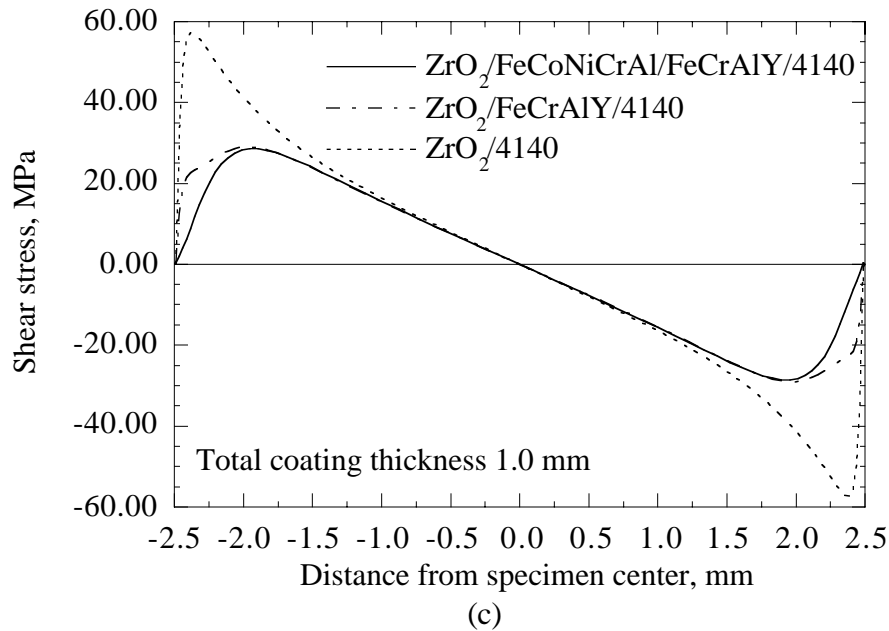


Fig. 8 (Continued) FEM stress distributions in thermal barrier coating systems under the condition of uniform temperature variation from 25 to 700°C. (a) Normal and shear stress distributions of a coating system without bond coats; (b) Normal and shear stress distributions of a coating system with two layer bond coats; (c) Influence of the bond coat type on edge shear stress distributions in the ceramic coating at the ceramic/bond coat interface.

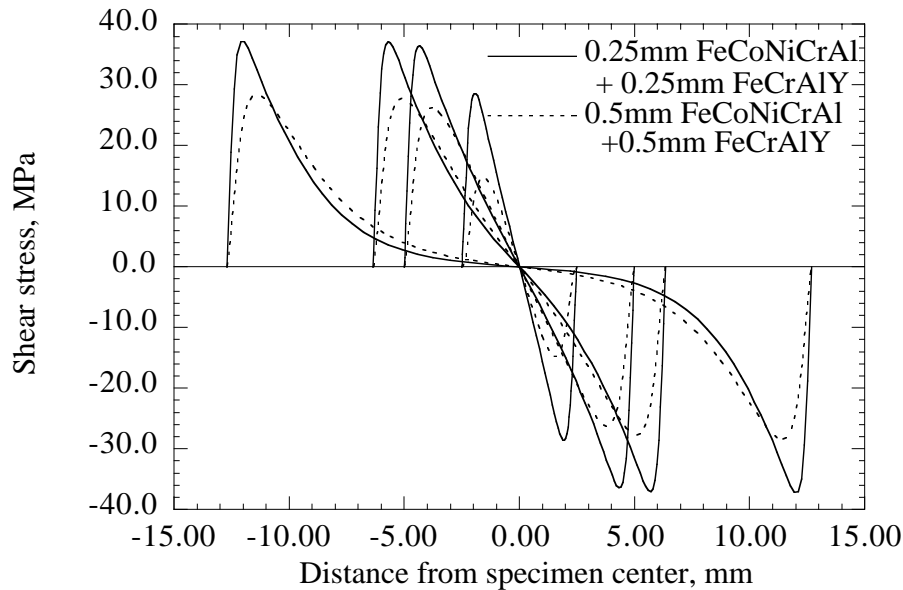


Fig. 9 Influence of bond coat thickness and specimen length on edge shear stress distributions in the ceramic coating at the ceramic/bond coat interface under the condition of uniform temperature variation from 25 to 700°C.

It has been realized that sintering-segmentation-enhanced delamination can be an important failure mechanism for a thermal barrier coating system during thermal cycling^[9]. The through-thickness cracks generated can create numerous edges in the ceramic coating systems. The modulus of the ceramic coating can also be significantly increased due to the sintering densification process. Figure 10 shows the shear stress distributions at the ceramic/bond coat and the bond coat/substrate interfaces for two effective ceramic modulus values (27.6 GPa and 100 GPa) to simulate the densification effect. It can be seen that the shear stresses at both interfaces increase with increasing the modulus of the ceramic coating. The increased stress concentrations due to sintering and oxidation of the bond coat and substrate can further accelerate the coating delamination process. The densification and oxidation processes in the coating systems increase the driving force for crack initiation and propagation, and reduce the coating adhesion. The graded bond coat systems proposed in this study can have beneficial effects in improving the coating performance.

In order to evaluate the stress concentrations in sintering segmentation ceramic coatings under thermal gradient heating conditions, FEM has been used to obtain the stress distributions of the segmented, through-thickness notch specimens described above. The stress distributions for a ceramic, two layer bond coat and 4140 substrate TBC system are illustrated in Figure 11. Since the notch width is wide enough, an edge stress concentration effect similar to the free edge case described above are expected near the ceramic/bond coat and the bond coat/substrate interfaces. However, under the present thermal gradient heating and low interfacial temperature conditions, a compressive (instead of a tensile) stress component σ_{yy} is developed. Therefore, in this case, shear induced cracking at the interfaces becomes a major mechanism for delamination crack initiation.

Experimental work has been performed to confirm several aspects of the coating edge effects predicted by the finite element analysis. The strain isolation of the bond coat system was studied by measuring the displacements in the ceramic coating attached to the substrate with various bond coat systems under the condition of uniform temperature variation from 25 to 700°C, and compared with the FEM calculations. The results are shown in Figure 12. Figure 12 (a) shows the effect of bond coat type on strain isolation. A lower thermal expansion bond coat gives a better strain isolation effect. It should be mentioned that although the one-layer FeCoCrNiAl bond coat appears to have the best strain isolation for the ceramic coating, it can generate significantly higher stresses at the bond coat/steel substrate interface as compared with the other bond coat systems. Figure 12 (b) shows the effect of bond coat thickness on strain isolation. The improved strain isolation effect by a thicker, multilayer, graded bond coat system suggest a reduced stress concentration at the ceramic/bond coat interface. Although discrepancies are observed between the experimental data and the FEM calculations, the strain isolation effects of the various bond coats obtained by both methods are similar and consistent. It should be noted that the FEM solutions did not consider any interfacial sliding and substrate plasticity or the possibility of low coating modulus near the interface regions.

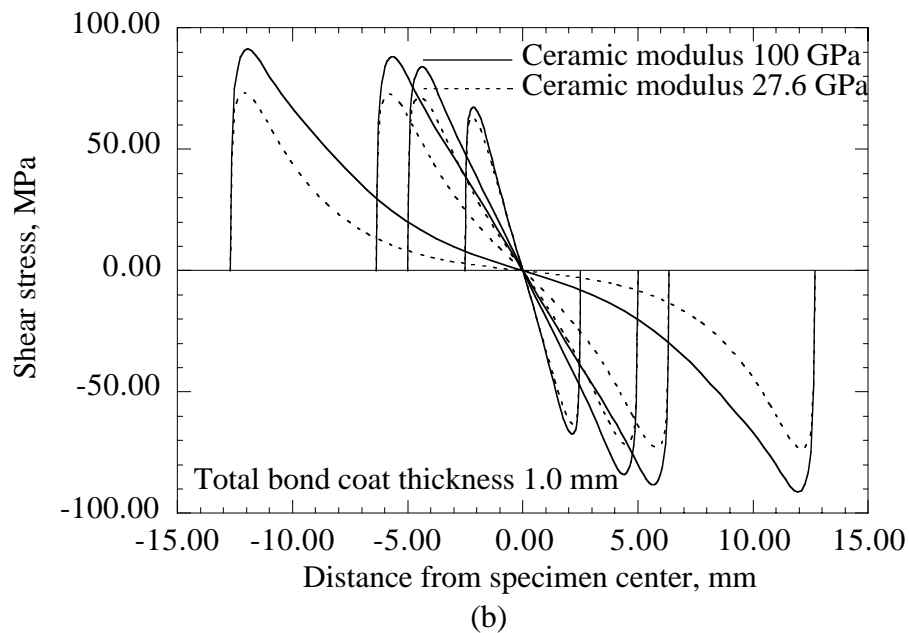
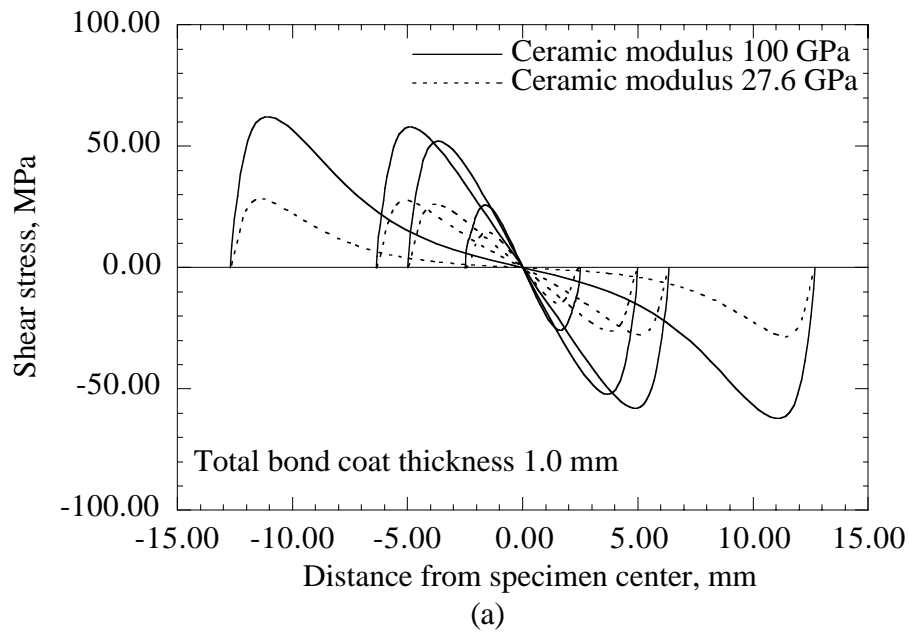


Fig. 10 Influence of increased ceramic elastic modulus on shear stress distributions in the ceramic/FeCoNiCrAlY/FeCrAlY/4140 system under the condition of uniform temperature variation from 25 to 700°C. (a) At the ceramic/bond coat interface; (b) At the bond coat/substrate interface.

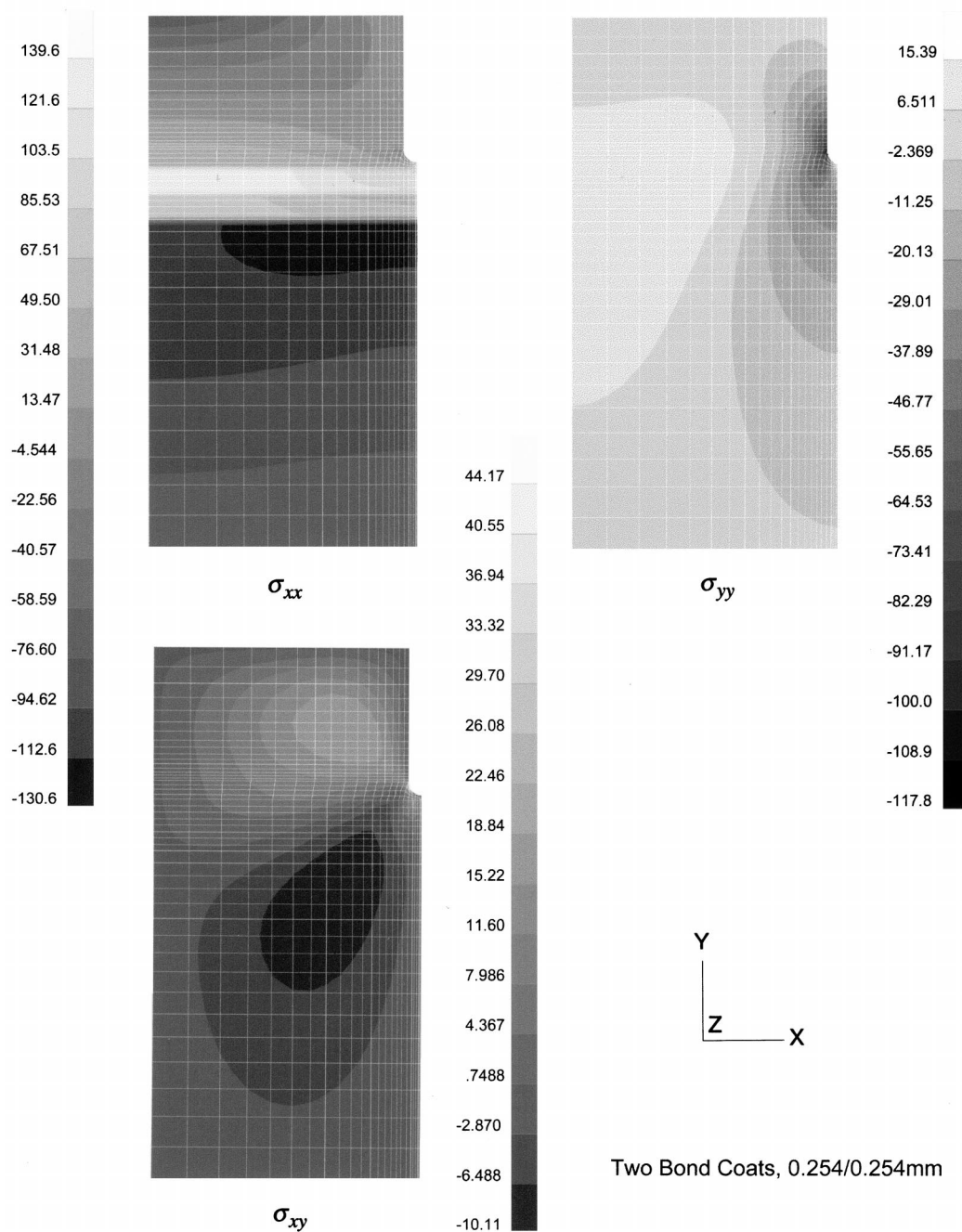


Fig. 11 FEM stress distributions near a notch edge for a ceramic, two layer bond coat and 4140 substrate TBC system under thermal gradient conditions. Ceramic surface temperature and 4140 substrate backside temperature are 850°C and 400°C, respectively.

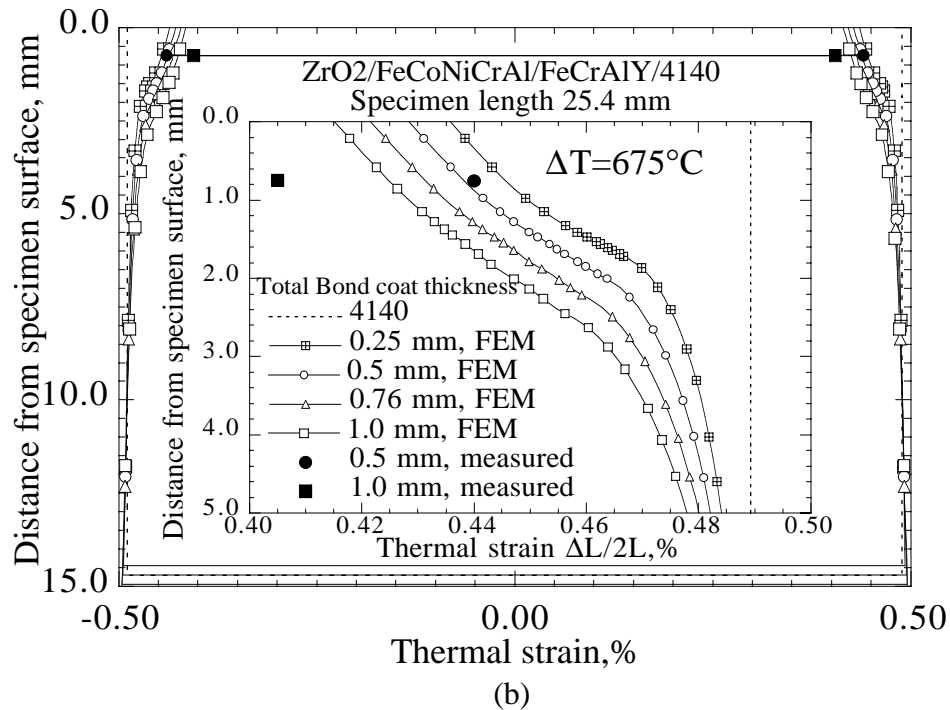
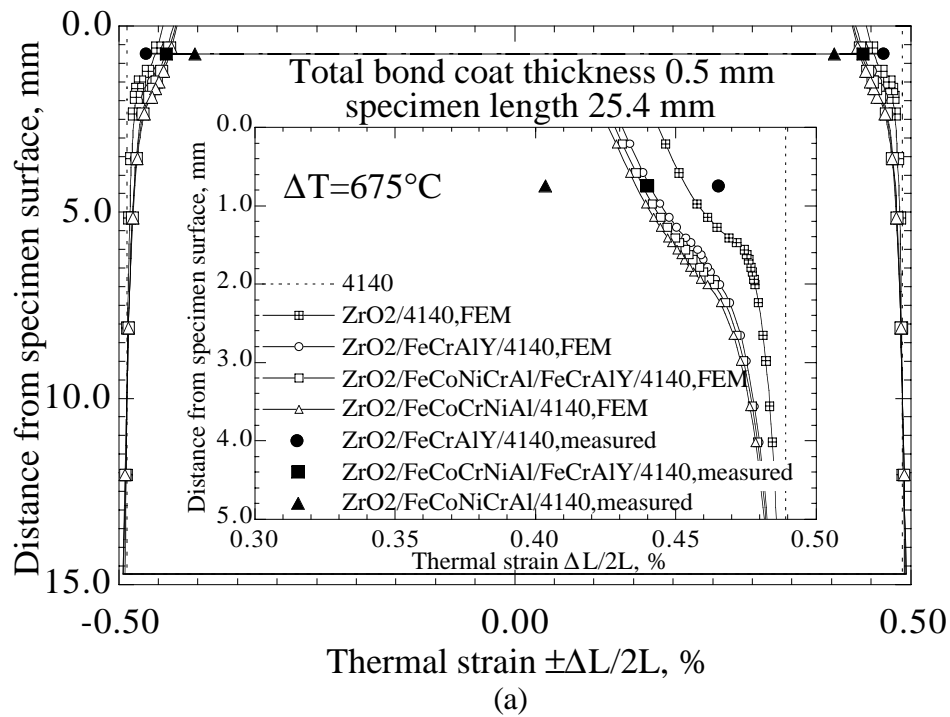
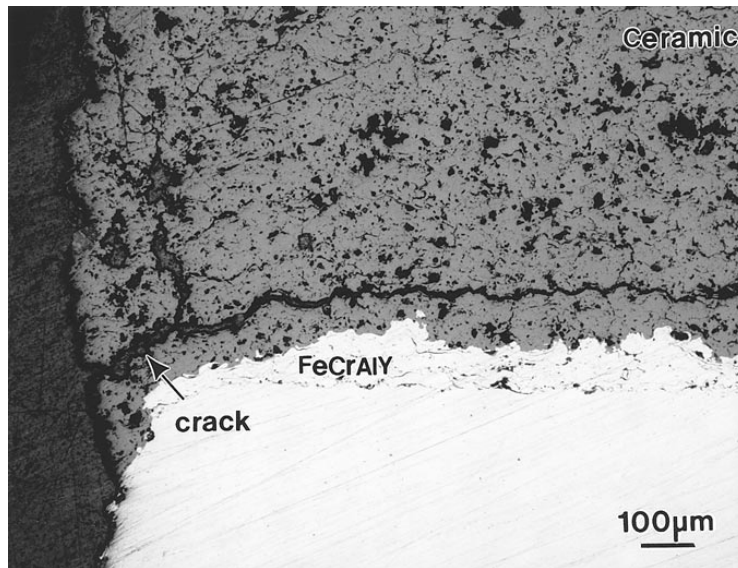


Fig. 12 Determination of strain isolation effect by dilatometry and FEM for thermal barrier coating systems under the condition of uniform temperature variation from 25 to 700°C (specimen center as the reference point). (a) The effect of bond coat type on strain isolation; (b) The effect of bond coat thickness on strain isolation.

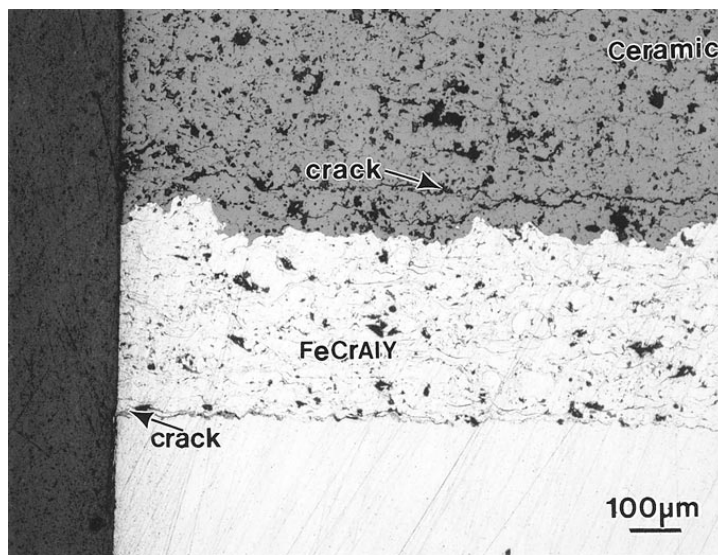
Laser thermal cycling experiments of segmented thermal barrier systems have also demonstrated that cracks are always initiated at the coating edges. As shown in Figure 13 (a), for a thin single layer FeCrAlY bond coat, the crack initiated and propagated in the ceramic coatings near the ceramic/bond coat interface. However, as shown in Figure 13 (b), for a thick single layer FeCrAlY coating, delaminations were observed at both the ceramic/bond coat and the bond coat/substrate interfaces. This is because the increase in FeCrAlY bond coat thickness can effectively shift the high stress concentration region from the ceramic/bond coat interface to the bond coat/substrate interface. For the segmented specimens, the long delamination cracks were observed near the through-thickness machined notch edges for the single layer bond coat systems, as shown in Figure 14 (a). The two-layer graded bond coat system in Figure 14 (b) showed a much better performance in resisting edge crack initiation and propagation under thermal cycling conditions at both the ceramic/bond coat and the bond coat/substrate interfaces, due to the improved bond coat strain isolation at both interfaces.

V. CONCLUDING REMARKS

A low thermal expansion and layer-graded bond coat system, that consists of plasma-sprayed FeCoNiCrAl and FeCrAlY coatings and a high velocity oxyfuel (HVOF) sprayed FeCrAlY coating, was developed for minimizing the thermal stresses and providing excellent oxidation resistance. Experimental results and finite element analysis show that the layer-graded bond coat system possesses lower interfacial stresses, better strain isolation and oxidation resistance in the thermal barrier coating system, which will lead to improved coating performance and durability.

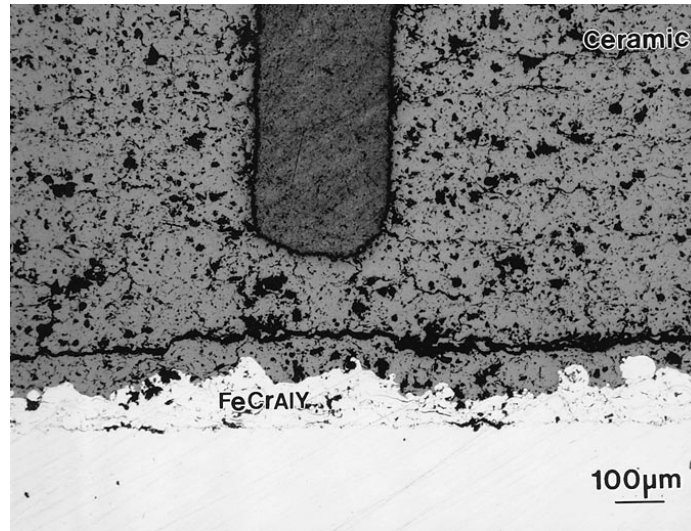


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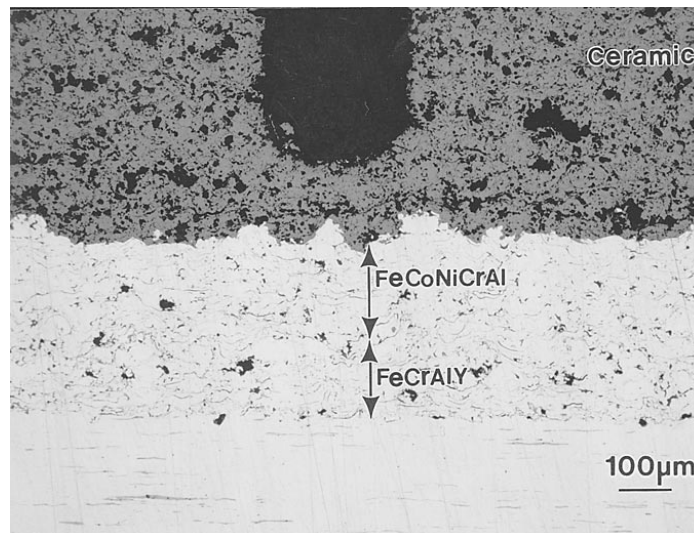


(b)

Fig. 13 Edge crack initiation and propagation in single layer FeCrAlY bond coat thermal barrier coating systems after 50 laser thermal cycles. (a) Bond coat thickness 0.127 mm; (b) Bond coat thickness 0.5 mm.



(a)



(b)

Fig. 14 Delamination crack initiation and propagation near the vertical machined notch edges for the thermal barrier coating systems after 50 laser thermal gradient cycles. (a) Single layer FeCrAlY bond coat thickness 0.127 mm; (b) Two-layer FeCoNiCrAl - FeCrAlY bond coat total bond coat thickness 0.5 mm (each layer 0.25 mm).

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